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TITLE: LOW TEMPERATURE PROPERTIES OF $CeCu_5$ AND RELATED COMPOUNDS

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LOW TEMPERATURE PROPERTIES OF $CeCu_5$ AND RELATED COMPOUNDS

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INTRODUCTION

The reported phase diagram[1] of the Ce-Cu system contains five compounds and a meta-stable sixth compound, $CeCu_{5.6}$.[2] $CeCu_6$ is by far the most investigated compound, primarily because of its heavy Fermion behavior at low temperature.[3] It is this last which makes comparisons with other Ce-Cu compounds of special interest. Several studies have been reported on both $CeCu_2$[5,6] and $CeCu_5$.[7,8] We present here our results on $CeCu_5$, partly because they include properties not included in Refs. 7 and 8, and partly because our results differ in one important respect, namely in the observation of two distinct phase transitions at low temperature.

Our samples of $CeCu_5$ were prepared by arc-melting the elements in argon atmosphere, followed by annealing at 700°C. It can be seen from the Ce-Cu phase diagram that contamination by the phase $CeCu_4$ and $CeCu_6$ is to be carefully guarded against. Both metallography and the specific heat results presented below indicate the absence of these additional phases in our samples. $CeAlCu_4$ samples were not annealed. $CeZn_3Cu_2$ and $CeAlZn_2Cu_2$ were prepared in sealed tubes.

The electrical resistivity behavior of $CeCu_5$ is that of a good metal, and one observes a resistance anomaly associated with a phase transition, most likely of magnetic origin, just below 4 K. The small size of this anomaly is indicative of the weak coupling between the Ce 4f-electrons and the conduction electrons in this compound. The temperature dependence of the resistivity here is in marked contrast, we note, to the Kondo-like behavior seen for $CeCu_6$.

We find a high temperature variation of the magnetic susceptibility corresponding to the full Ce $4f^1$ moment of 2.54 μ_B. Our results below 10 K (Fig. 1) show an antiferromagnetic anomaly at 3.8 K, with a second feature at 3.6 K. It is the presence of this double anomaly that does not appear in previous reports. Magnetization measurements in a variety of fields (Fig. 2) allow us further to prepare a magnetic phase diagram of the two magnetic features, also shown in Fig. 3.

The conclusion that two phase transitions are involved is further supported by thermal expansion data (Fig. 4), where two anomalies are seen. (The magnetostriction is also included in Fig. 4.) The positions of the features in thermal expansion relative to the magnetic anomaly have been used in refining the magnetic phase diagram in Fig. 3. It is not

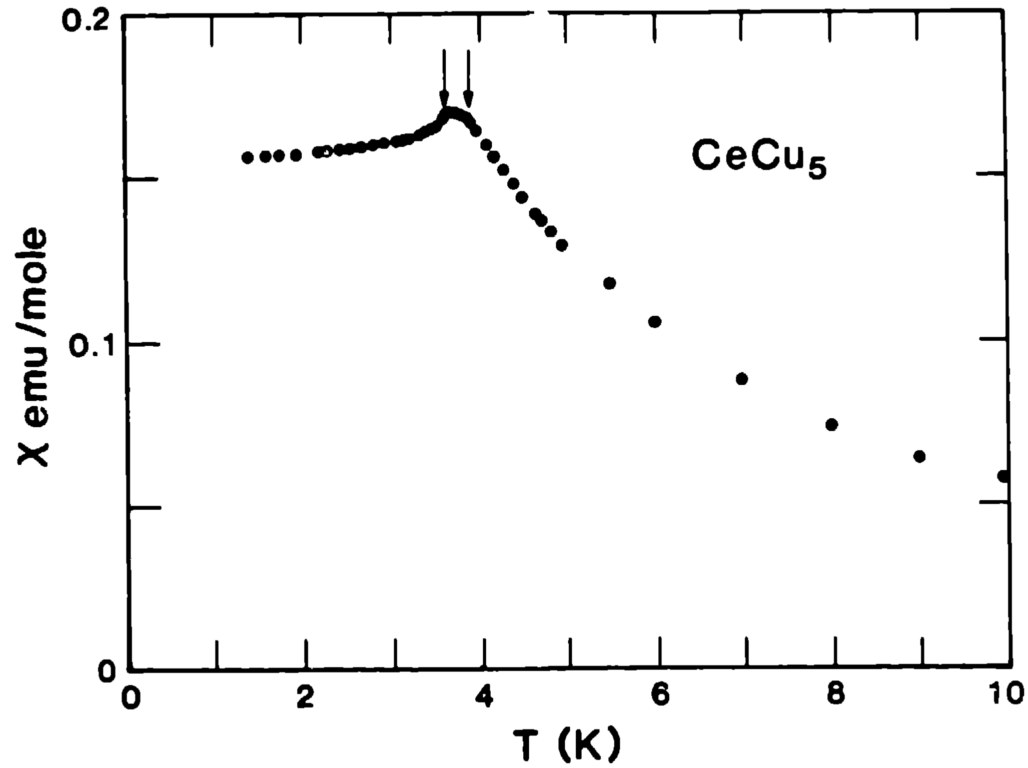

Fig. 1. Low temperature magnetic susceptibility of $CeCu_5$. The arrows mark the two phase transitions.

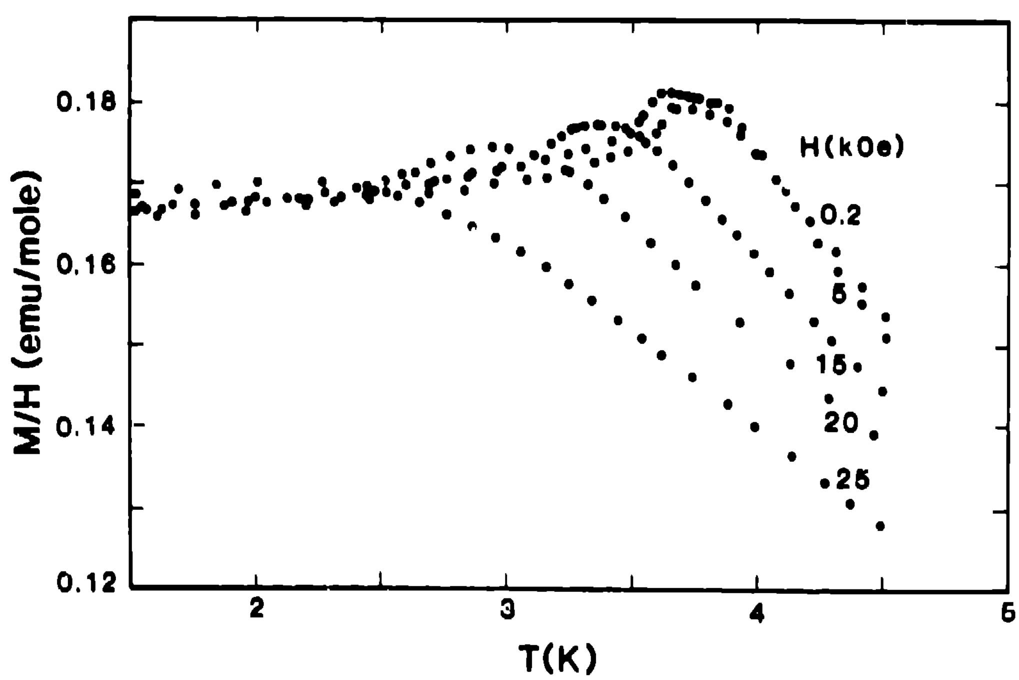

Fig. 2. Temperature dependent M/H data for $CuCu_5$ in various fields.

clear exactly which part of a feature in the thermal expansion corresponds, for example, to the upper transition seen in the magnetic susceptibility data, but there are clearly two distinct signatures in the expansion data. It is interesting that the amplitude of the upper signature rapidly enlarges in field (Fig. 5), while staying nearly at the same temperature, while that of the lower signature rapidly diminishes and moves to lower temperature.

The two anomalies are also clearly present in the specific heat data (Fig. 6). The first point to make is that the two peaks correspond closely to the minima in the thermal-expansion coefficient. An entropy integration of the double transition yields approximatley $R\ell n2$, corresponding to the involvement of a single crystal-field doublet of the Ce 4f configuration. It is further found that the low-temperature extrapolated electronic specific heat $\gamma \simeq 50$ mJ/mole-Ce K^2, a value somewhat enhanced above that of a simmple metal.

In order to clarify that the presence of two phase transitions was not due to trace $CeCu_4$ contamination, we performed specific heat measurements on $CeCu_4$. We were unable to prepare completely single phase material here, some $CeCu_5$ always being apparent via a specific heat anomaly near 3.8 K. Using the amplitude of this anomaly as a measure of the amount of $CeCu_5$ present as second phase in the $CeCu_4$ sample, we could estimate the low temperature specific heat properties of $CeCu_4$. We find a γ of approximately 400 mJ/mole-Ce K^2, placing it in the medium heavy Fermion category. This large γ for $CeCu_4$ sets an upper limit of ~1/8 for $CeCu_4$ contamination of our $CeCu_5$ sample, and, at this level, nothing seen in the specific heat of the $CeCu_4$ sample near 4 K could account for the second transition.

An unexpected feature of the $CeCu_5$ behavior is the presence of two phase transitions in a magnetic system consisting, as it appears, of a

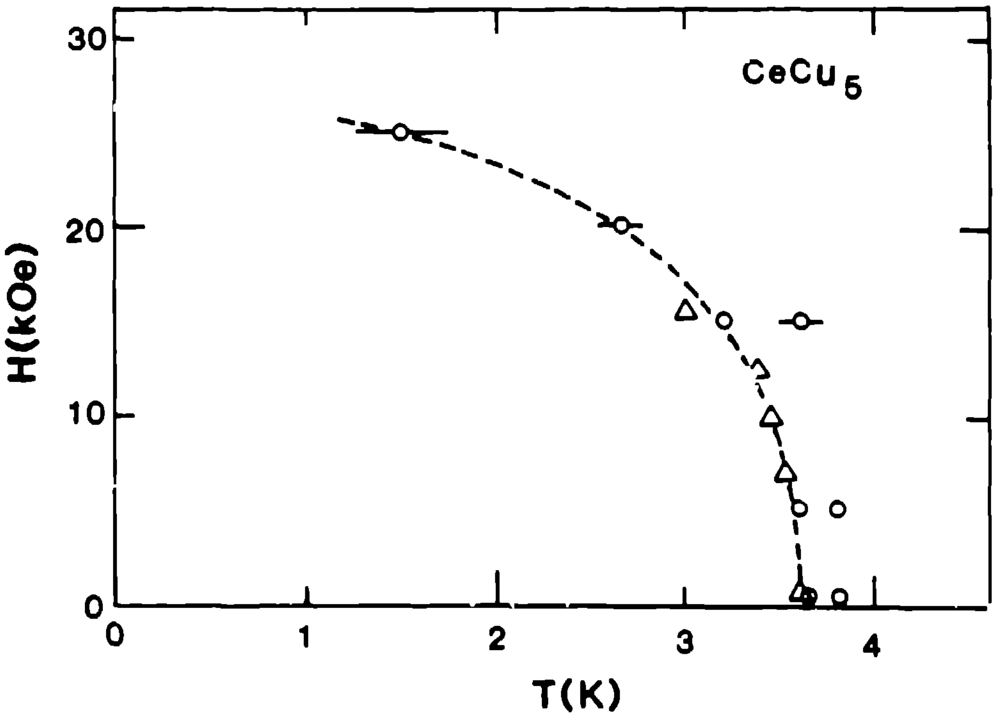

Fig. 3. H-T phase diagram of $CeCu_5$.

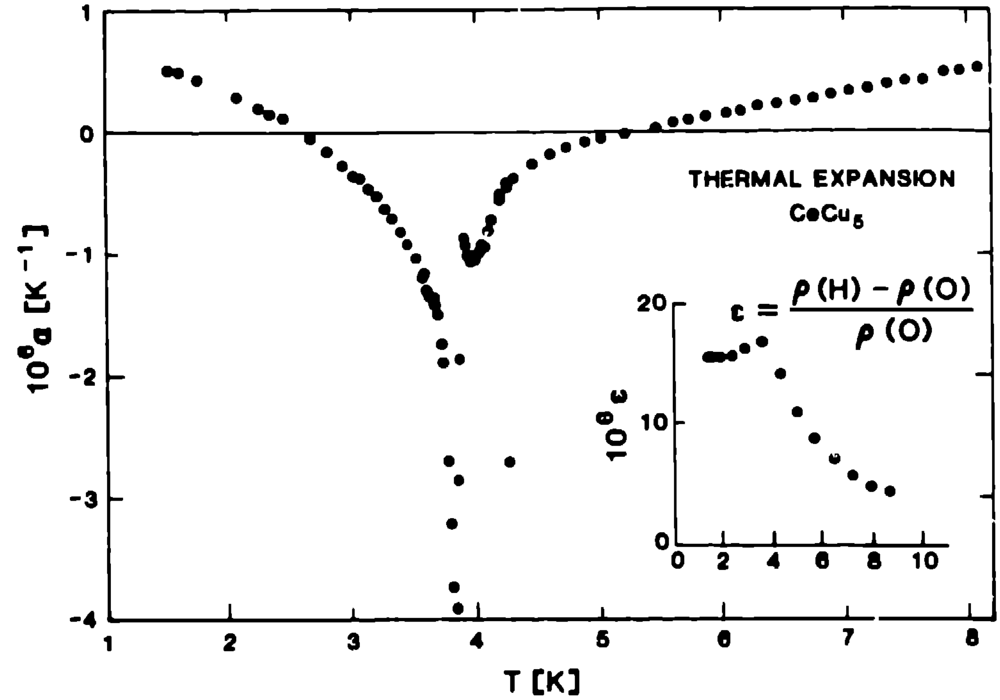

Fig. 4. Thermal expansion of $CeCu_5$. Inset shows the temperature dependence of the magneto-striction.

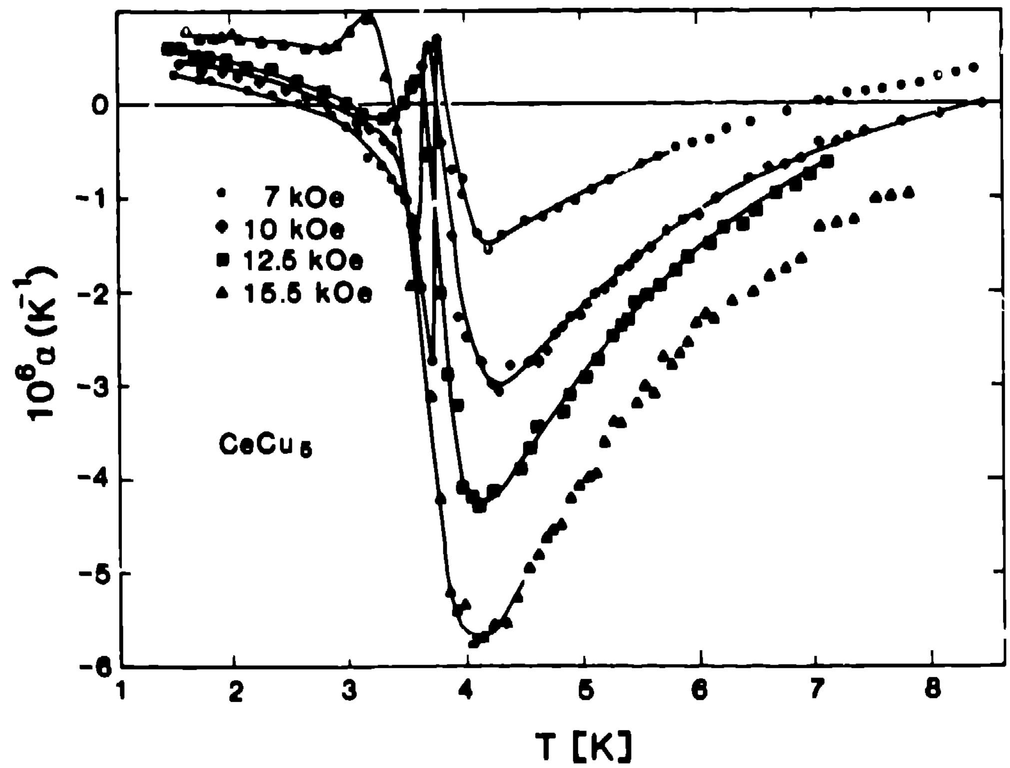

Fig. 5. Magnetic field dependence of thermal expansion of $CeCu_5$.

crystal field ground state doublet. It is also somewhat surprising to find this compound sandwiched between two heavy Fermion compounds in the binary phase diagram.

The hexagonal compound $CeCu_5$ contains two inequivalent Cu sites, a two-fold and a three-fold site. Separate utilization of these sites is found in the isostructural $CeRh_3B_2$. Takeshita et al.[9] have shown that Al can be substituted for Cu into the three-fold site in $CeCu_5$. We have made measurements on both $CeAlCu_4$ and $CeAl_2Cu_3$. The most interesting of these is $CeAlCu_4$, for which we show specific heat data to low temperature in Figs. 7 and 8. We find an enormously enhanced value of γ of about 2 J/mole-Ce K^2 below 1 K, and no magnetic ordering. γ has already fallen off in $CeAl_2Cu_3$ to a much lower value (~540 mJ/mole K^2 at 1.6 K). Additionally, a sample of composition $CeGaCu_4$ was found to order at 0.7 K from a high γ state.

We have also found it possible to prepare $CeZn_3Cu_2$ in this crystal structure. This material orders magnetically at 6 K with an entropy integrated to T_N of approximately $R\ell n2$ (Fig. 9), similar to $CeCu_5$, but with only a single phase transition. We have also measured the specific heat of $CeAlZn_2Cu_2$ to 1.6 K and found a very large value of C/T. ac-susceptibility measurements of this compound to 0.4 K show no sign of magnetic ordering.

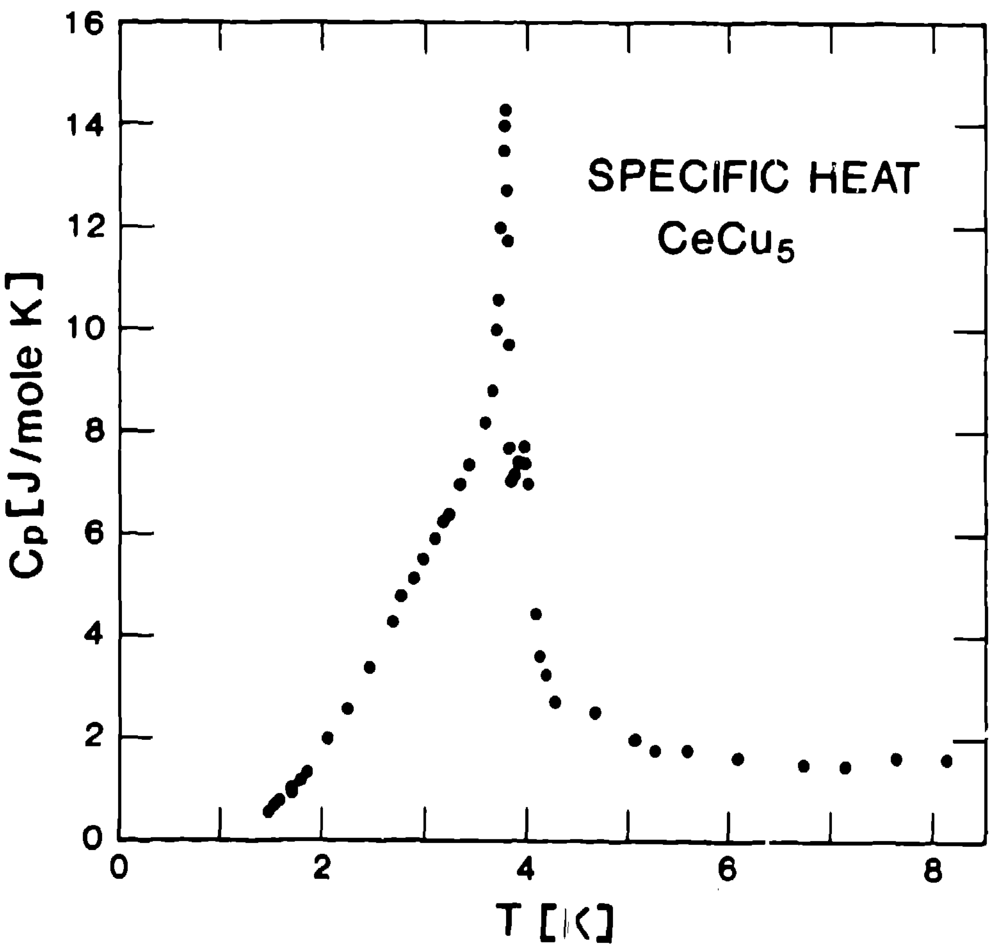

Fig. 6. Low temperature specific heat of $CeCu_5$.

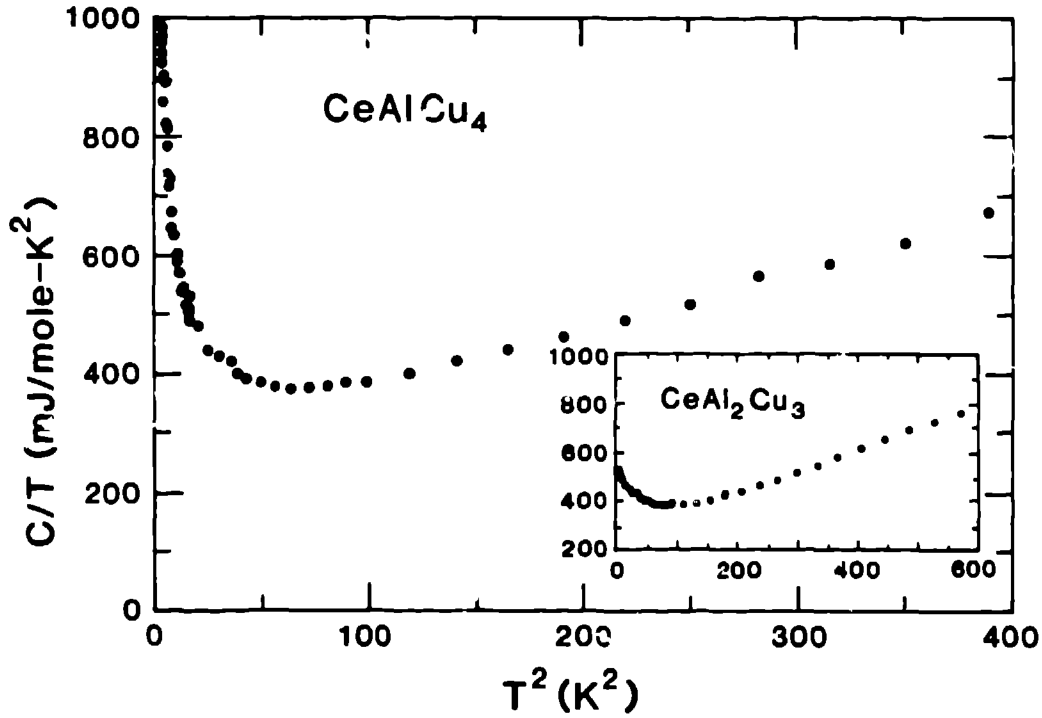

Fig. 7. C/T for CeAlCu$_4$ and CeAl$_2$Cu$_3$ (inset) versus T^2 at low temperature.

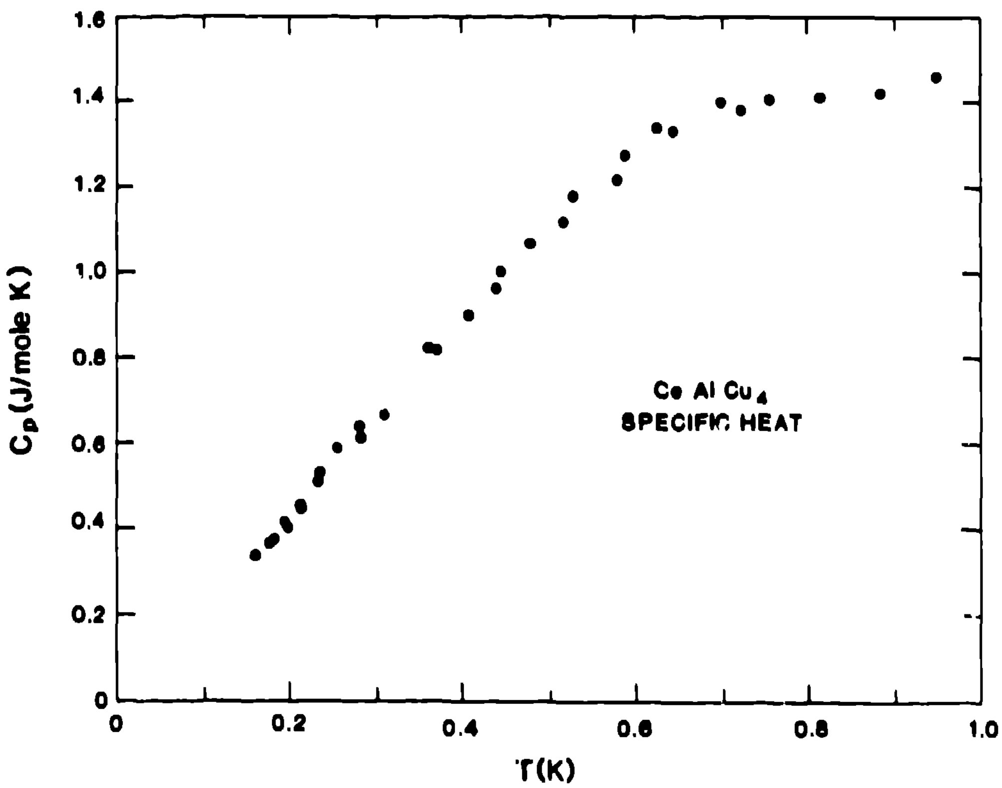

Fig. 8. Specific heat below 1 K for CeAlCu$_4$.

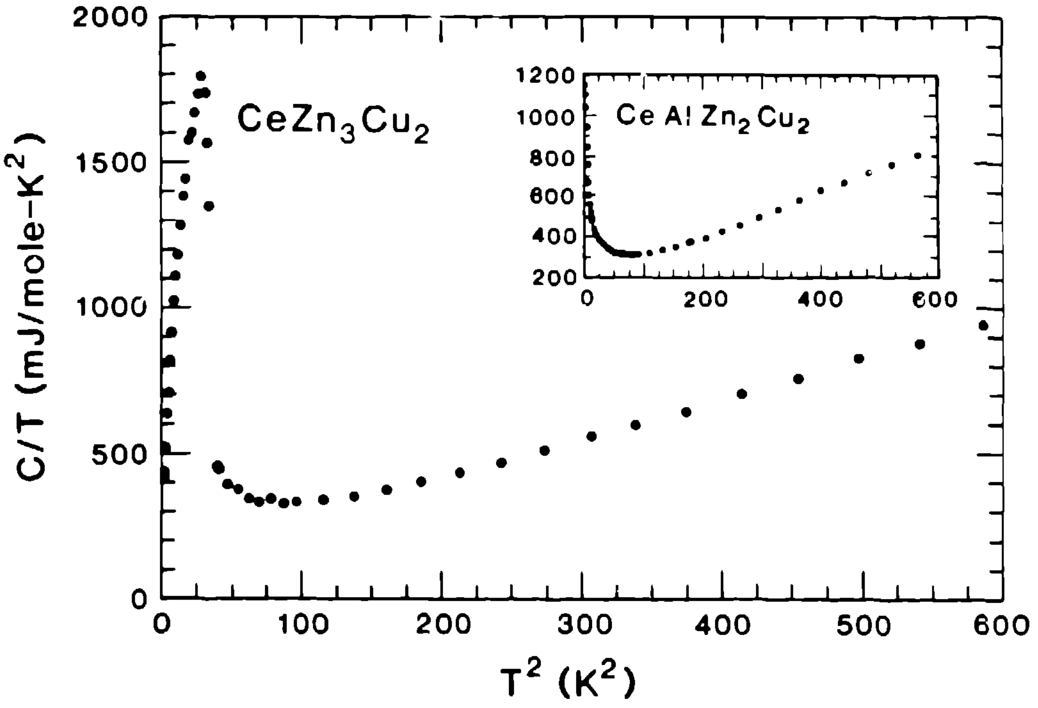

Fig. 9. C/T for $CeZn_3Cu_2$ and $CeAlZn_2Cu_2$ (inset) versus T^2.

It is remarkable that Al substitutions in both $CeCu_5$ and $CeZn_3Cu_2$ have such large effects. Because the electronic structures of $CeCu_5$ and $CeZn_3Cu_2$ must be quite different,we believe this points to a special effect of the Ce-Al interaction that results in these very large γ values at low temperature in the Al substituted compounds.

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